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From the inside out: Upscaling organic residue analyses of archaeological ceramics

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ABSTRACT

The investigation of organic residues associated with archaeological pottery using modern analytical chemical methods began in the 1970s. It was recognised early on that the analysis of lipids (i.e. fats, waxes and resins) preserved in surface residues or the fabric of single potsherds, representative of single vessels, was a powerful method for ascertaining pottery use, with a high degree of specificity. Subsequent developments saw a significant change in scale, with studies often involving lipid analyses of tens to hundreds of potsherds per archaeological assemblage, providing information that extended beyond pottery use. The identification of animal and plant foodstuffs processed in pots provides insights into herding and farming, and can also detect trade in exotic organic goods. Information about the environment and climate can be extrapolated from the isotopic composition of compounds detected in potsherds, potentially providing novel avenues of investigation. The direct dating of lipids in potsherds is opening up new opportunities for building archaeological chronologies, while the integration of lipid residue analyses with other environmental and cultural proxies within interdisciplinary projects is already providing unprecedented insights into past lifestyles, from site to regional scales.

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1. Introduction

The maxim 'absence of evidence is not evidence of absence' is often invoked in archaeology, and holds especially true for organic remains, which generally degrade over archaeological timescales. Developments in organic residue analysis have been largely driven by developments in analytical chemistry, particularly advances in chromatographic and mass spectrometric instrumentation. The key advantage of modern analytical methods is their capacity to resolve the many biomolecular components that typically comprise organic residues in archaeology. Furthermore, the high sensitivities of the instruments are especially compatible with the extremely low concentrations of organic components preserved. Determination of the structures of compounds, or suite(s) of compounds, originating from plant and animal sources, known as 'biomarkers' (Philp and Oung, 1988), in sediments, ceramics and other matrices, substantially widens the available evidence base for archaeology (Evershed, 2008b).

Visible organic residues are well-known to archaeologists, appearing as encrusted deposits adhering to the interior or exterior surface of a vessel, and could derive from burnt residues (Fig. 1a), soot, etc., deposited by heating of the vessel over fire, or from materials used as decoration, sealants or adhesives (Fig. 1b). However, despite being well known, these visible residues are not common in pottery

assemblages and are prone to post-burial and post-excavation loss during cleaning of sherds and/or contamination (Fig. 2). Since avoiding sherd or removing soil/handling contamination from visible or surface residues is so fraught with difficulties, such residues are not the preferred choice for this type of analysis. Instead, the majority of analyses now target absorbed organic residues preserved in unglazed ceramic vessels, which generally originate from the original contents that were either stored or processed in the vessels, representing either a single use or an accumulation of cooking events over a vessel's lifetime.

Early attempts to extract and analyse absorbed residues revealed the presence of highly unstable compounds, such as unsaturated fatty acids, and human-derived lipids such as cholesterol (Rottländer, 1990), probably arising from modern contamination. Thorough mechanical removal of potsherd surfaces before powdering the sub-surface fabric allows contaminating compounds from the burial environment and/or handling to be removed (Condamine et al., 1976; Heron et al., 1991). Critically, migration of lipids from the soil to the buried potsherd has been shown to be negligible (Heron et al., 1991). A sampling protocol involving the removal of potentially contaminated surface layers is now an accepted approach, and has been applied to thousands of sherds to date. The approach appears to be applicable to both freshly excavated sherds and those from museum collections. Experience has shown that sub-sampling between 2 and 3 g of clean potsherd yields sufficient lipid to work with, while still providing a reasonable area of the vessel wall to overcome local heterogeneity factors. Traditionally lipids have been

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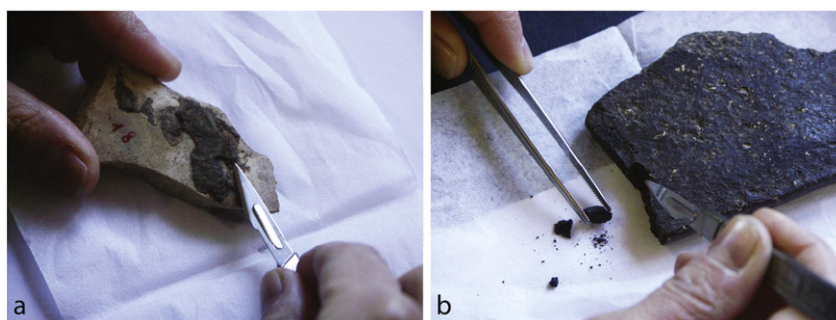


Fig. 1. Sampling of visible residues adhering to sherds and interpreted as (a) burnt food crust and (b) an adhesive used in pottery repair.

extracted from visible residues and powdered fabric using organic solvent mixtures (Evershed, 2008a; Evershed et al., 1990). However, a more recent protocol involving the direct hydrolysis and methylation of lipids allows both high-throughput and higher recoveries of lipids from archaeological potsherds to be achieved (Correa-Ascencio and Evershed, 2014). The loss of compositional information when complex lipids such as triacylglycerols or wax esters are hydrolysed is the only disadvantage of this method, although sherds can often be re-examined using solvent extraction if needed (Correa-Ascencio and Evershed, 2014).

The high sensitivities of instrumental methods such as gas chromatography and mass spectrometry allow microgram (μg) to nanogram (ng) amounts of compounds to be detected and identified (Fig. 3). Even higher sensitivity (picograms; pg) can be achieved using selected ion monitoring (SIM) methods, for example for the detection of specific marine biomarkers (Cramp and Evershed, 2013; Evershed et al., 2008a). The advent of gas chromatography–combustion–isotope ratio mass spectrometry (GC-C-IRMS) in the 1990s introduced the possibility of accessing stable isotope information from individual biomarker structures, opening up a range of new avenues for the application of organic

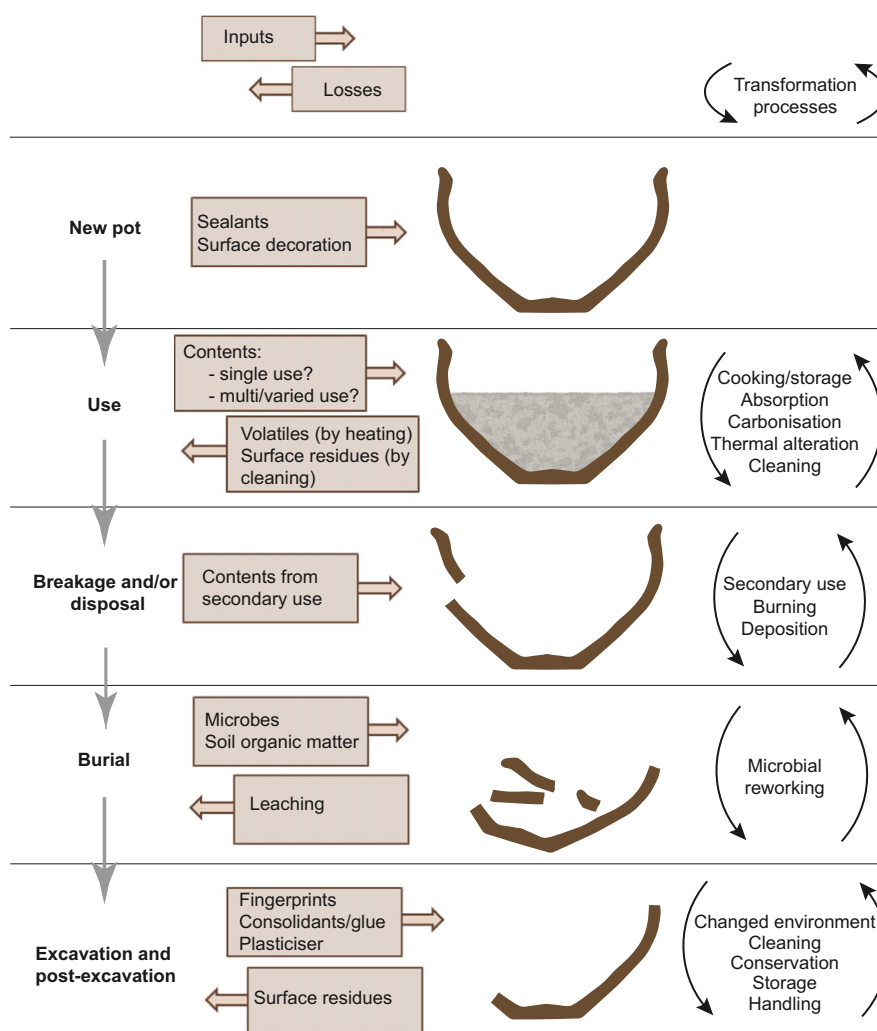


Fig. 2. Main inputs, losses and transformation processes affecting the survival and composition of visible and absorbed organic residues in archaeological ceramics. Adapted from Historic England (2017b) after Stacey (2009).

residue analysis in archaeology (Evershed et al., 1994, 1997a). The advent of analytical methods, which allow amorphous and invisible organic materials from archaeological contexts to be detected and identified, has thus contributed significantly to answering hitherto intractable archaeological questions, across both temporal and spatial scales. This article provides a review of the contributions organic residue analysis has made across the themes of vessel technology, subsistence and foodways, movement and connectivity, palaeoenvironment and palaeoecology, and chronology.

2. Vessel technology

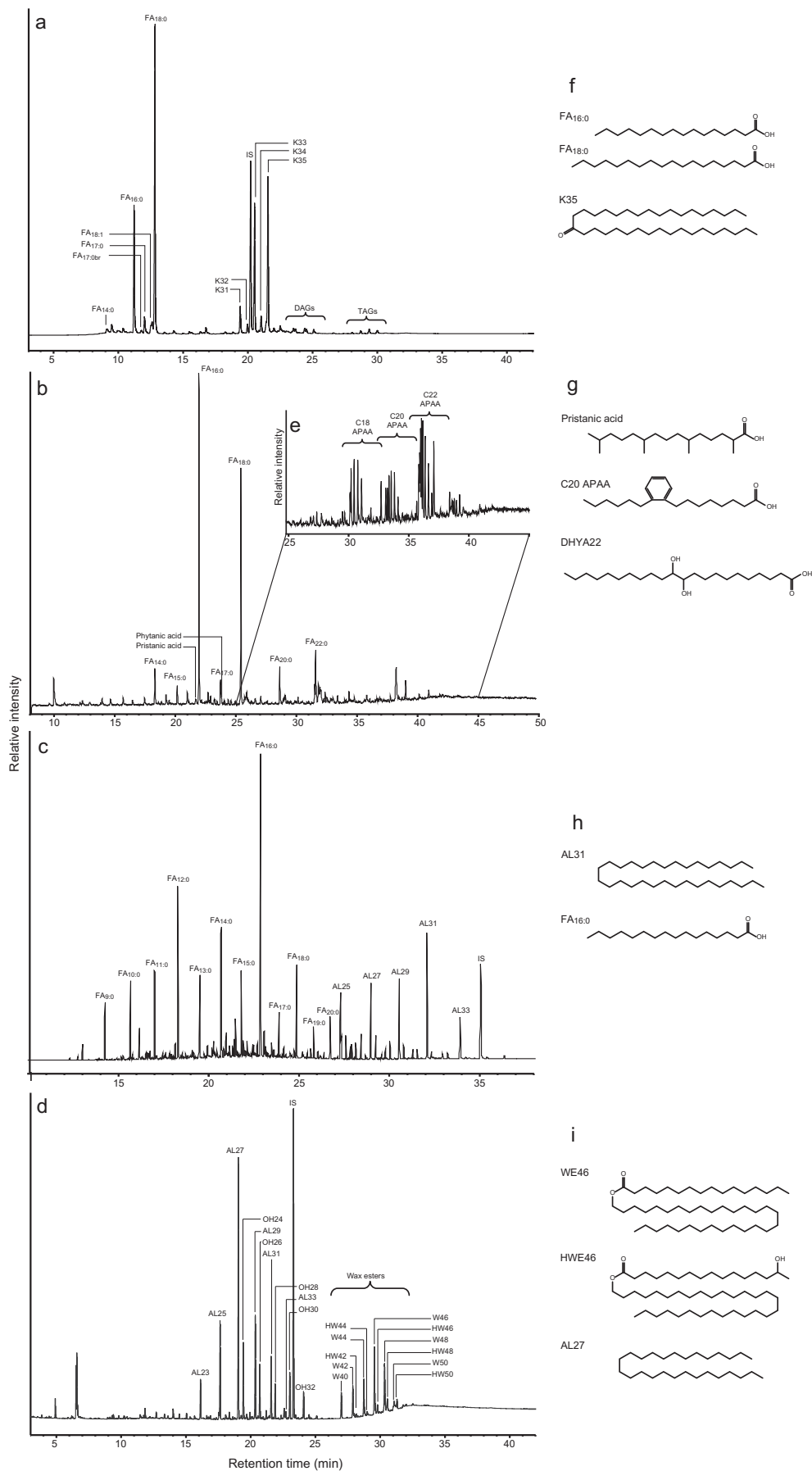
The analysis of organic residues from pottery has been highly effective in gaining insights into a range of different aspects relating to ceramics, including production, use, repair and technological change and specialisation. Technologies involved in the production of ceramic vessels that can be identified using organic residue analysis include manufacture (including sealing), decoration and repair. For example, unglazed fabrics offer the highest potential for the retention and survival of absorbed residues, yet their surfaces need to be sealed to decrease the permeability of the fabric and make them effective containers for liquids. To date, a range of sealants such as waxes, resins and bituminous materials have been identified on archaeological ceramics, dating back to the Neolithic. A known sealant, beeswax, largely undetectable by the naked eye, has been identified as a water-proofing agent on Early Neolithic collared flasks (Fig. 3d; Poland; 5400–4800 BCE; Salque et al., 2013) and Bronze Age east Mediterranean Red Lustrous Wheel made ware (16th century BCE–early 12th century BCE; Knappett et al., 2005). Another commodity used as a sealant was birch bark tar, used to line the interior walls of handled jugs at the Late Neolithic site of Makriyalos (Greece; 5400–4900 BCE; Urem-Kotsou et al., 2002) and to seal vessels at the Iron Age site of Grand Aunay (France; 3rd–1st centuries BCE; Regert et al., 2003). Sometimes sealants are preserved as surface deposits, for example black/brown sticky deposits found on four 6th-century BCE vessel sherds from Naukratis (Egypt) were identified as pitch derived from conifer wood, possibly used to line the interior of the vessels (Stacey et al., 2010). Excavations at Anuradhapura (Sri Lanka) identified a number of buff ware vessels lined with bitumen, known as ‘torpedo jars’. Dated stylistically to between the 3rd and 9th centuries CE, it is thought that the coating was used to seal the permeable containers so that they could be used to transport liquid commodities such as oils, perfumes or wine. The bitumen was sourced to Susa in Iran through biomarker distributions and isotopic signatures, suggesting the existence of long-distance trade relationships between Sri Lanka and the Middle East (Stern et al., 2008).

An example of applied decoration is found on bitumen-painted ceramics from Late Neolithic Tell Sabi Abyad (northern Syria; c. 6100 BCE; Connan et al., 2004). Significantly, the bitumen used to paint the ceramics probably originated from two different areas in northern Iraq, suggesting long-distance trade and exchange networks existed at that time. The adhesive used to repair a Roman Ecton ware jar at West Cotton (Northampton, UK) was identified as birch bark tar (CE 43–10; Charters et al., 1993), and bitumen, identified from the distribution of sterane and terpane biomarkers, was used to repair bowls at Tall-e Abu Chizan (south-western Iran), a late prehistoric (Middle Susiana to Middle Uruk) settlement dating from 5000 to 3900 BCE (Connan et al., 2008). Bitumen had a long history of use at Kavaşan Höyük (Turkey), from the 14th to the 4th century BCE, being used mainly as a water-proofing agent on pottery (on both the inner and outer vessel walls) but also as a glue to repair broken Kavaşan ceramics. Thick bituminous crusts found on some jars may also represent the storage and processing of bitumen on-site (Connan et al., 2013). Significantly, it seems that the bitumen originated from the Eruh outcrops, situated 120 km east of the site, and was used at Kavaşan Höyük during the occupation span of the

site, implying the existence of a long-established trade route (Connan et al., 2013).

Qualitative and quantitative evaluation of lipid residues can demonstrate different methods of food technology, for example boiling or roasting, associated with particular commodities or vessel types. Significantly, information about the use of vessels as cooking pots can be derived from the presence of ketones (Fig. 3a). These ketones, when present with odd carbon number distributions of $C_{29:0}$ to $C_{35:0}$ (with $C_{31:0}$, $C_{33:0}$ and $C_{35:0}$ being particularly abundant), originate from the pyrolysis of acyl lipids and ketonic decarboxylation reactions, which occur in unglazed ceramic vessels, possibly during cooking, when the temperature exceeds 300 °C, and are thought to accumulate gradually with repeated use (Evershed et al., 1995; Raven et al., 1997). In some vessels, these ketones only appear to form in certain areas, providing clues about how the pots were being heated. For example, jars from some Romano-British sites bear distinctive patterning in ketone distribution (Faverdale, UK; 2nd century CE; Cramp et al., 2012). The ketones are concentrated around the rims of cooking jars, indicating that the vessels were used for boiling. During boiling, the lower parts of vessels are cooled down by the evaporation of water through the ceramic porous walls; the upper part of vessels reach higher temperatures, which is conducive for the formation of ketones.

Relationships between form and function can be examined usefully with organic residue analysis, as the fabrication and style of vessels are often related to the commodities processed within them. Vessel capacity, as well as shape, are likely to relate very closely to different potential functions for the various styles of pottery (e.g. Rice, 2006). Narrow, high-mouthed jars and jugs were probably used to store and handle liquids, whereas broader bowl-shaped pots might have been used to cook solid food. For example, absorbed lipid residues were extracted from three categories of vessels, ‘saucepan pots’, jars and bowls, from the British Iron Age sites (8th century BCE–1st century CE) of Maiden Castle, Danebury Hillfort, Yarnnton Cresswell Field and Stanwick, to determine what commodities were being processed in each container and also to establish whether any of these particular vessel types were consistently used to process specific commodities. The study revealed that at sites where ‘saucepan pots’ predominated, milk products were processed in those vessels, while at sites where jars predominated, then these latter vessel types were preferentially associated with dairy products (Copley et al., 2005c). A study of perforated potsherds dating from 5200 to 4800 BCE and excavated from sites occupied by the first central European farmers (Linearbandkeramik; LBK) was undertaken by Salque et al. (2013), to test the hypothesis that such sieves were used as cheese-strainers (Bogucki, 1984). The identification of dairy fats, in conjunction with the specific (sieve) vessel shape, provided compelling evidence for prehistoric cheese-making. Furthermore, the presence of dairy residues in bowls suggested their use in combination with the sieves in the cheese-making process, as seen in other archaeological contexts (Gouin, 1990). At the same sites, cooking pots were used to process ruminant meat, while collared flasks were waterproofed with beeswax, probably before being used for handling low-lipid content liquids. Significantly, this specificity of food-stuffs detected in pots from the region of Kuyavia (Poland) has provided the first evidence for Early Neolithic specialisation in pottery use (5400–4800 BCE; Salque et al., 2013). Questions of form and function were also addressed in organic residue analyses of two distinct medieval vessel types (lamps and ‘dripping dishes’) from the Causeway Lane excavation, Leicester (UK; 11th–14th centuries BCE). Compound-specific analyses of the $C_{16:0}$ and $C_{18:0}$ fatty acids demonstrated the presence of ruminant animal fat, for example sheep, goat or cattle (tallow), in the lamps and non-ruminant animal fat, for example pig, in the ‘dripping dishes’, demonstrating specialisation of vessel use (Mottram et al., 1999). Specialisation in pottery use was also evident at the Mexican city of Teotihuacan around 200–550 CE, where bacterial markers for an alcoholic beverage produced from the fermented sap of *Agave* plants (pulque) were only detected in amphorae and ollas.



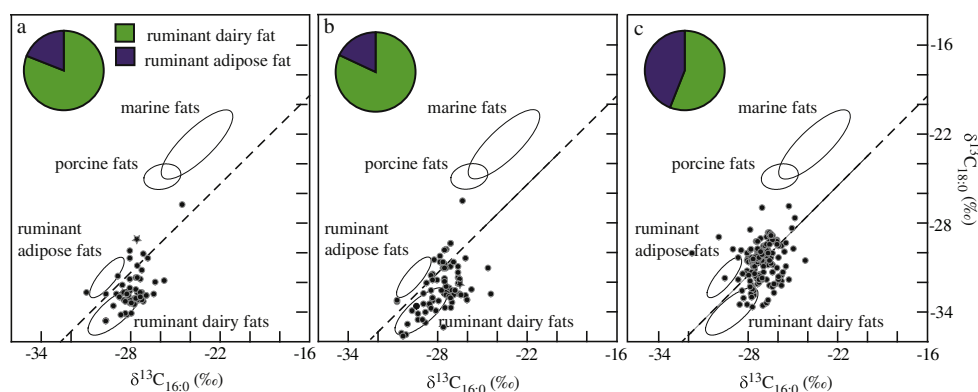


Fig. 4. $\delta^{13}\text{C}$ values of $\text{C}_{16:0}$ and $\text{C}_{18:0}$ fatty acids preserved in pottery from northern Britain, the Outer Hebrides and the Northern Isles of Scotland, dating to the (a) Early (pre-4600–3700 BCE), (b) Middle (3600–2900 BCE) and (c) Late (2900–2300 BCE) Neolithic; star symbol indicates where aquatic biomarkers were also detected. Adapted from Cramp et al. (2014b).

The presence of pine resin in the vessels, probably used as a post-firing waterproofing agent, suggests that they were used for storing or processing liquids (Correa-Ascencio et al., 2014).

3. Subsistence and foodways

The area of archaeological research where organic residue analysis has proven most informative is subsistence and foodways, from the broad scales of society and geography to, more recently, the narrower scales of community and household (Evershed, 2008b). Organic residue analysis has contributed to our understanding of the invention and adoption of pottery technology and its effect on changing subsistence strategies and foodways during the Neolithic and Bronze Age.

For example, organic residue analyses of early pots from Britain and the western Baltic have revealed a consistent difference in the contents of Neolithic pottery between the two regions. The analysis of >400 pottery sherds from 48 Early to Late Neolithic assemblages (pre-4600 BCE to 2300 BCE) across the British Isles by Cramp et al. (2014b) revealed compelling evidence for a substantial dairy economy established immediately and widely upon the advent of the Neolithic in Britain (Fig. 4). The almost ubiquitous detection of saturated carboxylic acids, and $\delta^{13}\text{C}$ values of fatty acids consistent with ruminant adipose and dairy fats, suggested a widespread focus of subsistence and foodways on domesticated livestock. Biomarkers for aquatic resources, such as long-chain vicinal dihydroxy acids and ω -(*o*-alkylphenyl)alkanoic acids (APAAs; Hansel et al., 2004; Hansel and Evershed, 2009), were detected in only two sherds (Fig. 3b). This is in contrast to the preceding Mesolithic period, which did focus on the use of aquatic species, as evidenced by numerous remains of shell middens along the Atlantic seaboard of Europe between Denmark and Portugal (Gutiérrez-Zugasti et al., 2011) and the stable carbon isotope analysis of human bone collagen from individuals either side of the Mesolithic–Neolithic transition in Britain (10950–6450 BCE; Richards et al., 2003). Conversely, analysis of 133 pottery sherds from the western Baltic, comprising pre-agricultural (Mesolithic) Ertebølle (5200–4000 BCE) and agricultural (Neolithic) Funnel Beaker wares (4100–2700 BCE) from coastal and inland sites, revealed that around 24% of Mesolithic sherds and 16% of Neolithic sherds from coastal sites contained isoprenoid fatty acids and APAAs, denoting aquatic resource exploitation. This suggests a more gradual change in subsistence and

foodways upon the advent of the Neolithic in this region (Craig et al., 2011).

These studies illustrate how organic residue analysis of early pottery has helped raise awareness of the non-uniformity of Neolithic culture across Europe, probably influenced by the diverse ecological and cultural environments inhabited by prehistoric peoples. In addition to large-scale or diachronic questions of past human diet, organic residue analysis offers the opportunity to investigate the cultural role of foodways and subsistence. The role of ceramic vessels in the manifestation of ideological or cosmological belief systems cannot be understated; the entire chaîne opératoire involved in obtaining, processing, consuming and disposing of food in itself contains both implicit and explicit aspects of society and culture (e.g. Bray, 2003; Dietler and Hayden, 2001; Farb and Armelagos, 1980; Goody, 1982; Klein and Murcott, 2014). For example, the strong association of dairy products and Carinated Bowls, recovered from a wide range of Early Neolithic sites across Britain and Ireland (pre-4600–3700 BCE; Copley et al., 2005c; Cramp et al., 2014b; Smyth and Evershed, 2016), marks the transition to an agricultural lifestyle, with the growing of crops and management of animals such as cattle, sheep and goats. This period marks one of the most profound transformations in human and animal relationships, with significant social, economic and ecological implications. Alongside this, the absence of marine biomarkers, suggesting that aquatic products were not processed in vessels, is hypothesised to indicate a broader ‘taboo’ or cultural rejection of food from the sea (Richards and Schulting, 2006; Thomas, 2003).

From the later Neolithic period, the study of a large number of organic residues from pottery indicates a strong correlation between probable ceremonial sites, pig-feasting and Grooved Ware pottery (Mukherjee et al., 2007, 2008). Later Neolithic Grooved Ware is thought to have originated in northern Scotland before 3000 BCE, subsequently spreading across Britain and Ireland over a period of approximately 1000 years, being used alongside other contemporary pottery styles. In southern Britain in particular, the Durrington Walls sub-style of Grooved Ware is commonly found associated with henges and timber circles, and sites with Grooved Ware in general seem to display a high prevalence of pig bones in their faunal assemblages. The investigation of ancient animal fats preserved in 385 vessels, comprising Grooved Ware and non-Grooved Ware from northern and southern Britain, revealed that Grooved Ware was used most intensively for processing

Fig. 3. Partial gas chromatograms of total lipid extracts characteristic of (a) animal fats, (b) aquatic resources (note that the DHYAs are not presented), (c) plant oils and (d) beeswax (adapted from Salque et al., 2013). (e) Mass chromatogram (m/z 105, 290, 318 and 346) showing APAAs in aquatic resources (same extract as b). (f–i) Examples of main biomarkers for the identification of the same foodstuffs. IS, internal standard (*n*-tetratriacontane); $\text{FA}_{n,i}$ fatty acids with *n* carbon atoms and *i* unsaturations; K, mid-chain ketones with 31–35 carbon atoms; DAGs, diacylglycerols; TAGs, triacylglycerols; Cn APAAs, ω -(*o*-alkylphenyl)alkanoic acids with *n* carbon atoms; DHYA, dihydroxy fatty acids; AL, *n*-alkanes; OH, *n*-alcohols; HW, hydroxy monoesters; W, wax monoesters.

porcine products in southern Britain (Fig. 5; Mukherjee et al., 2007, 2008). This association contrasts with both contemporaneous non-Grooved Ware and Scottish Grooved Ware, confirming a special, probably ritualistic, role of Grooved Ware for pig-feasting, and moreover indicating the development of regional ideological usages for pottery types. Interestingly, Craig et al. (2015) recently studied a large assemblage of Grooved Ware vessels from Durrington Walls to investigate intra-site variation in the use and deposition of pottery. Ruminant products seemed to be preferentially processed in the ceramics, although pig products were consistently detected in pots from pit features. Dairy residues were associated with ceremonial spaces, suggesting differences in consumption practices between public and more private domestic spaces.

4. Movement and connectivity

Detecting chemical traces of plants and animals in pottery sherds in locations they do not naturally inhabit can serve as a powerful proxy indicator for the mobility of past populations, networks of exchange and trade, and the pervasiveness of ritual beliefs and cultural practice. In southern Finland, acidic soils result in the exceptionally poor survival of archaeological remains, with animal bone normally encountered as small cremated fragments. Such poor preservation has left researchers unable to examine systematically when domesticates were first introduced to areas above the 60th parallel north and with what regularity farming was practised in this challenging environment. However, molecular and carbon stable isotope analyses on absorbed residues across a chronological sequence of pottery types has revealed a striking shift in commodity use (Cramp et al., 2014a). The Comb Ware of 4th millennium BCE hunter–fisher–forager groups contained lipid residues predominantly or exclusively of marine origin, while the southern Finnish Corded Ware (c. 2500 BCE) displayed values typical of terrestrial

ruminant fats, half of which were milk fats, which must have originated from domesticated stock. Residues from the later Kiukainen Ware (c. 2000 BCE) appear to have contained both marine and terrestrial resources, indicating subsequent modification of subsistence strategies in the region. The strong contrast between Comb Ware and Corded Ware use, only detectable through organic residue analysis, supports the hypothesis that the latter pottery type represents the successful introduction of novel subsistence practices into Finland. This demonstrates that organic residue analysis can itself act as a proxy where other lines of evidence are not present. For instance, in Ireland and many parts of south-western and northern England, acidic soils lead to poor preservation of faunal remains, precluding interpretations of animal husbandry practices in prehistory.

While the introduction and exploitation of domesticates in Ireland has been confidently assigned to the Early Neolithic (c. 3800 BCE), species identification is often extremely challenging and the construction of kill-off profiles using age-at-death data, from which the management of dairy herds is sometimes inferred (e.g. Payne, 1973; Vigne and Helmer, 2007), is almost impossible. A systematic programme of organic residue analysis (>450 pottery vessels from 15 Early, Middle and Late Neolithic sites; 4000–2500 BCE) has been invaluable in highlighting the antiquity and apparent embeddedness of a particular subsistence regime (Smyth and Evershed, 2015). Nearly 90% of the animal fats had $\delta^{13}\text{C}$ values indicative of milk fats. Indeed, ruminant milk fats were the dominant type of fat observed in residues from vessels dating to all phases of the Neolithic. As in Finland, domesticated animals and dairying can be linked to the appearance of pioneering farming groups. However, in Ireland this way of farming did not shift substantially during nearly a millennium and a half. The relative robustness of archaeological potsherds and the high-precision analytical techniques developed around them provides an unrivalled approach to tackling questions of longer term connectivity and social change.

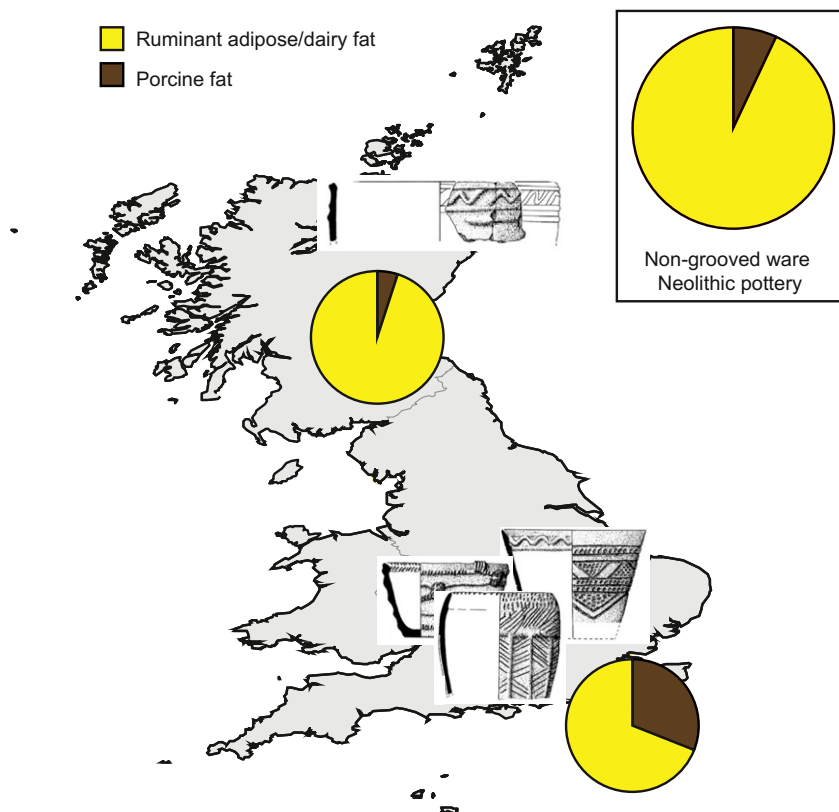


Fig. 5. Map showing examples of the northern and southern styles of Grooved Ware in Late Neolithic Britain (4th–3rd millennia BCE) and proportions of sherds containing predominantly porcine fats associated with northern and southern Grooved Ware and non-Grooved Ware pottery. (Data from Mukherjee et al., 2007, 2008).

Organic residue analyses can also provide insight into mobility and connectivity through the molecular analysis of resinous exudates from plants, particularly trees. Resins were extremely desirable commodities in the past, and indeed remain so, for their various aromatic, adhesive, antibacterial and aesthetic properties. Analytical techniques such as gas chromatography–mass spectrometry (GC–MS) and pyrolysis GC–MS (Py GC–MS) can identify characteristic biomarkers in degraded resins characteristic for certain plant species. As resin-yielding plant species are distributed unevenly across the globe, it is possible to narrow down or even isolate their place of origin (Langenheim, 2003; Mills and White, 1994; Regert et al., 2008, after Cordemoy, 1911), providing information on past exchange networks and the spread of cultural practices.

In recent work examining Romano-British (3rd–4th centuries CE) mortuary rites, organic residue analysis was undertaken successfully on both the residues visible on the bodies themselves and on the microscopic detritus found in the mortuary containers (Brettell et al., 2014, 2015). Suites of terpenoid biomarkers were detected in more than a quarter of the burials, representing three different plant families: coniferous *Pinaceae* resins, Mediterranean *Pistacia* spp. resins (mastic/terebinth) and *Boswellia* spp. gum-resins (frankincense/olibanum) from southern Arabia or beyond. Whether purchased in Britain or brought over by an immigrant family, the presence of these resins indicates considerable Roman cultural influence on this ostensibly ‘remote’ outpost of the Roman Empire. Similarly, systematic analysis of resin-like fragments from the medieval fortified warehouse of Sharma on the Yemeni Hadramawt coast is prompting a re-examination of the commercial network that developed throughout the Indian Ocean during the Middle Ages (11th century CE; Regert et al., 2008). The Hadramawt coast was renowned for its trade in frankincense, but molecular analyses have indicated that most of the fragments of resin discovered at Sharma were related to the exploitation of East African copal, with only two instances of frankincense detected. Indeed, interest in eastern African copal as an aromatic may have developed as early as the 7th or 8th century CE, as it has recently been detected, via GC–MS, on a brass artefact thought to be part of an incense burner from Unguja Ukuu on Zanzibar, one of the earliest known trading ports in coastal East Africa (Crowther et al., 2015). Molecular analyses are also providing information on the European markets for this medieval incense trade, and how it evolved following its partial collapse with the spread of Christianity. GC–MS analysis has securely identified a series of perforated domestic pots as incense burners, which appear in southern Belgian graves during the 11th and 12th centuries CE (Baeten et al., 2014). Alongside the precious and exotic frankincense, the analyses detected traces of aromatics such as juniper and possibly pine tar, suggesting that the expensive frankincense may have been ‘cut’ with cheaper, more local incense ‘ingredients’.

5. Palaeoenvironment and palaeoecology

The most common commodity detected in potsherds is animal fats, as identified by the presence of two main saturated fatty acids, palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acids. The foods that animals eat exhibit characteristic isotopic signatures (Gannes et al., 1997) and isotopic analyses ($\delta^{13}C$) of fatty acids extracted from archaeological potsherds are therefore a reflection of the diet consumed by the animals and thus can provide information about the environment from which they came (Copley et al., 2003; Mukherjee et al., 2005). The analyses of modern reference fats collected from non-ruminant (meat, e.g. from pigs) and ruminant (meat and milk, e.g. from sheep, goat or cattle) animals demonstrates the wide range of $\delta^{13}C$ values observed in fatty acids as a result of inhabiting diverse isoscapes (Dunne et al., 2012). The presence of C_4 plants in the diet of animals causes an enrichment in ^{13}C (higher $\delta^{13}C$ values) in the fatty acids of the animal products (Dunne et al., 2012). Similarly, an enrichment of up to 3‰ is observed

in water-stressed plants (Farquhar et al., 1989) and could be detected in animal fats extracted from potsherds from the Near East (Evershed et al., 2008b). In the same way, wide ranges of $\delta^{13}C$ values observed in fatty acids extracted from pottery vessels from the site of Takarkori (Libyan Sahara; 8100–2600 BCE) have been interpreted as signifying diverse pastoral modes of subsistence. Vertical transhumance could have been practised between the mountainous site of Takarkori in the winter and summer pastures on the former lakes shores, generating a wide range of $\delta^{13}C$ values in the animal fats (Dunne et al., 2012). Verification of the isotopic evidence for vertical transhumance is confirmed by the archaeological record (di Lernia, 1999; di Lernia et al., 2013).

Animal products processed in pottery vessels can be connected directly with the archaeozoological assemblage at a site. Lipid residue analyses and archaeozoological studies thus provide complementary evidence regarding dietary practices and herding strategies. A study conducted by Evershed et al. (2008b), comprising the analyses of >2200 sherds from the Near East and south-eastern Europe (7000–3500 BCE), found that milk residues were detected in the highest numbers from sites where cattle were the predominant species in the faunal assemblage, implying that cattle were the primary producers of milk. Similarly, high abundances of pig skeletal remains at British Grooved ware sites (4th–3rd millennia BCE) occurring simultaneously with the processing of non-ruminant (porcine) fats in Grooved Ware vessels, demonstrates the correlation between foodstuffs and archaeozoological assemblages (Copley et al., 2005a; Mukherjee et al., 2008). The compelling correlation between lipid residue analyses and faunal assemblages suggests that lipid residue analyses can be used as a proxy for animal exploitation at sites where bones have not survived (Cramp et al., 2014a; Salque et al., 2012; Smyth and Evershed, 2016).

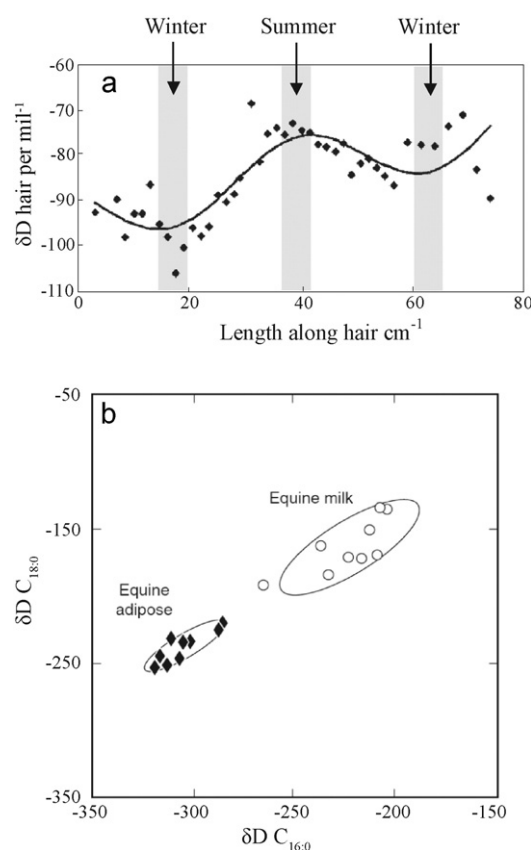


Fig. 6. (a) Seasonal variations in the stable hydrogen isotope composition of meteoric water are recorded in horse hair (adapted from Sharp et al., 2003). (b) δ^2H of the $C_{16:0}$ and $C_{18:0}$ fatty acids from modern horse adipose and milk fats. The confidence ellipses are mean \pm SD of the values exhibited by modern reference fats (Outram et al., 2009).

The construction of slaughtering profiles based on dental wear from cattle and sheep/goat mandibles excavated at archaeological sites provides insight into animal management and herding practices (e.g. Vigne and Helmer, 2007). A comparison between animal products processed in ceramic vessels and slaughtering profiles provides independent evidence for the exploitation of milk and carcass products at a site, as shown for the Early Neolithic site of Colle Santo Stefano (Italy; 5840–5460 BCE; Salque et al., 2012) and more generally in the Mediterranean region (Debono Spiteri et al., 2016). Despite an early attempt to characterise animal fats using the distributions of fatty

acids in triacylglycerols (Mirabaud et al., 2007), lipid residue analyses remain largely non-species specific. Archaeozoological studies thus provide the species-specificity lacking in the lipid residue analyses.

Identification of early horse domestication at the site of Botai in Kazakhstan (c. 3500 BCE) was made possible by the environmental signals that are recorded in lipid residues in the form of the hydrogen isotopic composition ($\delta^2\text{H}$ values) of fatty acids (Outram et al., 2009). As observed in the analyses of modern reference mare's milk and carcass fats, higher $\delta^2\text{H}$ values of fatty acids recorded in the seasonal (summer) production of mare's milk contrast with the lower $\delta^2\text{H}$ values

Table 1

Selected case studies involving the use of organic residue analyses. Adapted from Historic England (2017a).

Site level question	Site-level analysis	Fit within large-scale questions	Example (site specific and broader context, if applicable)
<i>Production, use, technological change and vessel specialisation</i>			
Manufacture, use, decoration and repair of vessels.	Encrusted and absorbed residues (as required)	Technological change, resource exploitation, trade relationships	a) Jōmon pottery (15,000–11,800 cal BCE) used for marine and freshwater fish processing. Earliest evidence for pottery manufacture and use (Craig et al., 2013). b) Birch bark tar used to repair Ecton ware jar at West Cotton, Northampton (Charters et al., 1993) c) Bitumen used as applied decoration on Late Neolithic ceramics from Tell Sabi Abyad, Syria (Connan et al., 2004). d) Beeswax as surface sealant (post-firing treatment) found on Bronze Age Mediterranean Red Lustrous Wheel ware (Knappett et al., 2005) and on Neolithic collared flasks (Salque et al., 2013).
Relationships between form and function (food and non-food products).	Comparative analysis of one or more defined vessel types (e.g. jars, dishes, bowls)	Tracing diversification and specialisation of vessel use through intra-, inter-site and regional comparisons. Assessment of broader trends in vessel use	a) British Iron Age 'saucepan pots', jars and bowls, to determine whether any of these particular vessel types were consistently used to process specific commodities. Milk products processed in 'saucepan pots' at sites where they predominated, yet at sites where jars dominated, they were preferentially associated with dairy products (Copley et al., 2005a, 2005b). b) Early Neolithic sieves, collared flasks, bowls and cooking pots from Kuyavia (Poland). Sieves used as cheese-strainers, bowls used in association with milk, collared flasks waterproofed with beeswax and cooking pots used to process ruminant carcass products (Salque et al., 2013). c) Roman mortaria from urban, rural and military settlements. High abundances of plant epicuticular waxes in the mortarium and (mainly) degraded ruminant carcass fats, suggesting that both animals and plant products were routinely processed in the same mortaria although it is not known if they were mixed together or processed on separate occasions (Cramp et al., 2011).
Vessel function (non-food).	Vessel/container fills (liquid or solidified)	Technological change, resource exploitation	Whitish cream preserved in a small Roman tin canister from London. Made from animal fat, starch and tin, likely of cosmetic or medicinal origin (Evershed et al., 2004).
	Assemblage of likely containers or matrices, e.g. lamps, candleholders	Technological change, resource exploitation	Visible residues adhering to metal candlesticks from 12th century Fountains Abbey. 'Recycled' beeswax from church candles was mixed with tallow to make domestic candles (Frith et al., 2004).
Spatial patterning/activity-specific – e.g. pottery associated with different structures or activities	Analysis or comparison of assemblages associated with particular context, e.g. industrial area, domestic settlement, cemetery, ceremonial centre	Ritual use of pots and feasting	Grooved ware was preferentially associated with pig consumption. Pottery from ceremonial sites associated with pig exploitation suggesting a ritualistic aspect to pork consumption (Mukherjee et al., 2008).
	Intra-site variation	Intra-, inter-site and regional comparisons to assess broader trends in vessel use	Intra-site variation in the use/deposition of pottery vessels detected at Bronze Age Trethellan Farm, where the 'ancillary' buildings were less likely than the 'residential' structures to contain potsherds yielding lipids (Copley et al., 2005d).
<i>Food preparation techniques</i>			
Food preparation techniques	Investigation of sherds from different parts of the vessel profile (e.g. base, middle and rim) from characterised vessel types	Technologies and cultural culinary practices, such as boiling and roasting	Jars from Romano-British sites with distinctive patterning in ketone distribution, likely arising from cooking practices (Cramp et al., 2012).

Table 1 (continued)

Site level question	Site-level analysis	Fit within large-scale questions	Example (site specific and broader context, if applicable)
<i>Diet, economy and subsistence</i>			
Diet, economy and subsistence	Assemblages of cooking pots (>30 vessels)	Large-scale subsistence shifts through time, e.g. introduction of farming, intensification of agriculture, episodes of invasion/immigration and cultural change.	a) Periods with limited evidence for subsistence, e.g. prehistory (rather than historic period). - Site level - variable abundance of carcass fats/dairy fats processed in vessels. - Broader scale - dairying at early Neolithic sites (Evershed et al., 2008b, Copley et al., 2005a, 2005b, 2005c, 2005d). b) Regions with poor animal bone preservation, specifically due to acidic soils. - Dairying in Neolithic Ireland. Acidic soils meant poor preservation of faunal remains, precluding interpretations of animal husbandry practices. Lipid analysis showed dairying was a significant component of the earliest farming practices in Neolithic Ireland (Smyth and Evershed, 2015, 2016) c) Regions encompassing various isoscapes. - Evidence for short-distance movement by the detection of animal fats with heterogenous isotopic composition, showing that these animals had varied diets (from mixed C ₃ /C ₄ to pure C ₄ diets, Dunne et al., 2012).
<i>Resource acquisition/exploitation, trade and exchange</i>			
Resource exploitation, trade and exchange	Analysis of amorphous masses of organic matter (e.g. lumps of 'resin')	Some commodities, such as resins, can sometimes be provenanced to their geographic origin, allowing reconstruction of trade/exchange routes	Direct evidence for the use of resinous exudates in mortuary rites in Roman Britain, providing information on practical and symbolic aspects of Roman mortuary rites. Coniferous resin, mastic/terebinth resin from the Mediterranean and frankincense from southern Arabia or eastern Africa, giving insights into Britain's relationship with the Roman Empire (Brettell et al., 2014, 2015). <i>Brassica oleracea</i> (cabbage) identified in Saxon/Medieval vessels at West Cotton, Northamptonshire (Evershed et al., 1991).
	Investigation of assemblages of likely containers or matrices	Diet and subsistence strategies, exploitation of resources, trade relationships	
<i>Dating of events (¹⁴C dating)</i>			
Timing of events (pottery use)	Radiocarbon dating of lipids extracted from potsherds.	Timing of change in practices, refining regional chronologies, dating reliably sites where organic materials (e.g. charcoal and bones) are absent from archaeological record.	Feasibility study on sherds from the dendrochronologically-dated Late Neolithic Sweet Track (Somerset Levels, UK; Berstan et al., 2008).

of horse meat fats, which integrate the year-round environmental signature (Fig. 6). The identification of lipids recovered from pots at Botai as originating from mare's milk, based upon $\delta^{13}\text{C}$ and $\delta^2\text{H}$ signatures, thus enabled the identification of early milking of horses. Combining the presence of milk residues in pots together with bit-wear visible on horse teeth, and a decrease in body size compared with wild horses, therefore provided compelling evidence for the early domestication of horses at Botai (Outram et al., 2009).

In addition to the palaeofauna, lipid residue analyses provide a method for investigating plant cultivation and exploitation in antiquity. For example, the detection of fatty acids in sherds from Qasr Ibrim (Egyptian Nubia; 1000 BCE–1800 CE) of similar molecular and isotopic composition as palm fruits (dates) provided direct evidence for fruit processing (Copley et al., 2001) and the use of different plant oils and animal fats as lamp fuels (Copley et al., 2005e). Plant biomarkers were also detected in sherds from Takarkori and Uan Afuda (Libya; 9th–7th millennia BCE), suggesting plant processing at the sites, and echoing the abundant desiccated plant remains excavated in the region (Dunne et al., 2016). The detection of plant biomarkers at high frequencies (60–90% of the residues) in Romano-British mortaria provided evidence for plant processing, underlining a shift towards new recipes, changing commodities and different cooking vessels in the Roman world between the 1st and 4th centuries CE (Cramp et al., 2011; Cramp and Evershed, 2015).

6. Chronology

Despite its obvious limitations, relative dating, based on the relative ordering of events through stratigraphies at individual sites, and typologies and seriations between sites, provides a powerful tool for building chronologies, particularly in instances where there is a dearth of organic material. However, the technique of radiocarbon (¹⁴C) dating, which provides dates that are both precise and accurate, has provided the vast majority of archaeological and palaeoenvironmental chronologies spanning the past 50,000 years (Bronk Ramsey, 2008). Absolute dating by radiocarbon is performed on samples containing organic or inorganic carbon, such as archaeological bones, wood or shells. Different sources of carbon are available in archaeological ceramics: (i) carbon present in clay with a radiocarbon age dependant on the clay source; (ii) carbon present in the temper, for example consisting of ground shells or vegetable matter, which could be contemporary with the manufacture of the pot; (iii) carbon derived from fuel in the kiln, with a date generally contemporaneous with the manufacture of the pot; (iv) carbon from food-stuffs accumulated during the use of pottery; and (v) carbon present in the burial environment (Gabasio and Evin, 1986). Despite problems such as contact with exogenous contaminants or sources of different ages, these sources of carbon have all been the object of dating analyses (De Atley, 1980; Evin et al., 1989; Gabasio and Evin, 1986; Hedges et al., 1992; Johnson et al., 1986; Messili et al., 2013; Taylor and Berger,

Table 2

Commodities that can be detected using organic residue analyses. The detection methods are as follows: GC–MS, gas chromatography–mass spectrometry; SIM, selected ion monitoring; GC–C–IRMS, gas chromatography–combustion–isotope ratio mass spectrometry; Py–GC–MS, pyrolysis coupled with gas chromatography–mass spectrometry; FTIR, Fourier transform infrared spectroscopy. Adapted from [Historic England \(2017a\)](#).

Source		Biomarkers	Specificity	Detection method	Complementary analyses	Examples
Terrestrial animal fats	Carcass fats	Saturated and monounsaturated fatty acids, $\delta^{13}\text{C}$ values of fatty acids.	Ruminant and non-ruminant animals carcass fats can be distinguished (using $\delta^{13}\text{C}$ values of fatty acids). No species-specificity (e.g. ruminants such as cattle, sheep or goat; Dudd and Evershed, 1998 , Copley et al., 2003).	GC–MS, GC–C–IRMS	Archaeozoological studies to provide species-specificity.	Ruminant animal fat (tallow), probably of sheep or goat, found in medieval lamps whilst non-ruminant animal fat, such as pig, present in ‘dripping dishes’ suggesting that the latter were probably used as receptacles for fat collection during spit-roasting (Mottram et al., 1999).
	Dairy fats	Saturated and monounsaturated fatty acids, including shorter-chain fatty acids, $\delta^{13}\text{C}$ values of fatty acids.	Dairy fats can be separated from carcass fats through $\delta^{13}\text{C}$ values of fatty acids ($\Delta^{13}\text{C}$ proxy; Copley et al., 2003). No species-specificity (e.g. ruminants such as cattle, sheep or goat; Dudd and Evershed, 1998 , Copley et al., 2003).	GC–MS, GC–C–IRMS	Archaeozoological studies (construction of kill-off profiles; Vigne and Helmer, 2007) to provide species-specificity (e.g. Outram et al., 2009).	Large-scale studies on sherds from the UK (Copley et al., 2005a, 2005b, 2005c, 2005d ; Cramp et al., 2014b), Ireland (Smyth and Evershed, 2015, 2016), Scandinavia (Cramp et al., 2014a ; Craig et al., 2011), the Near East (Evershed et al., 2008b), Africa (Dunne et al., 2012) provided evidence for the emergence of milk exploitation during the Neolithic in those regions.
Aquatic fats	Aquatic fish, shellfish and mammals	Vicinal dihydroxy acids (DHYAs; Hansel and Evershed, 2009), isoprenoid acids (IPAs; Hansel et al., 2004 , Copley et al., 2004) and ω -(<i>o</i> -alkylphenyl)alkanoic acids (APAAAs; Hansel et al., 2004), $\delta^{13}\text{C}$ values of fatty acids.	Species from marine/ocean-like ecosystems can usually be distinguished from freshwater based on $\delta^{13}\text{C}$ values of fatty acids. Marine ecosystems are complex and higher-level specificity would be challenging.	GC–MS (SIM), GC–C–IRMS	Bulk collagen $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analyses; archaeozoological studies (e.g. Cramp et al., 2014b).	Scale of aquatic resource exploitation at the onset of the Neolithic (Cramp et al., 2014b ; Craig et al., 2011). Earliest pots (Jōmon period, Japan, 16,950 to 13,750 cal BCE) used to process freshwater and marine organisms (Craig et al., 2013).
Plant	Oils	Saturated ($\text{C}_{16:0}$ and $\text{C}_{18:0}$) and unsaturated fatty acids e.g. $\text{C}_{18:1}$ and $\text{C}_{18:2}$, hydroxy fatty acids, dicarboxylic acids.	Very specific biomarkers may exist for some oils, e.g. radish oil (see example), although the majority of plant oil biomarkers only indicate the processing of oils generally.	GC–MS	Archaeobotanical studies	Radish and castor oil used as illuminants in archaeological lamps at Qasr Ibrim, Egypt (Copley et al., 2005e).
	Waxes	Wax esters, long-chain <i>n</i> -alkanes, <i>n</i> -alkanols and even-numbered long-chain fatty acids.	Can be very specific depending on source, e.g. biomarkers found for <i>Brassica</i> although the majority of plant waxes are not specific and can only indicate processing of leafy plants.	GC–MS		<i>Brassica oleracea</i> (probably cabbage) identified in Saxon/Medieval vessels at West Cotton, Northamptonshire (Evershed et al., 1991). Mixing of leafy plants and animal fats identified in Roman mortaria (Cramp et al., 2011)
	Resins (inc. fossil resins), tars and pitches	Terpenoids including sesqui-, di- and triterpenoids.	Resins can often be provenanced to the botanical family of origin, and sometimes even to genus, allowing their geographic origin to be pinpointed. Distillation of resins, which produces tars or pitches, can also be identified.	GC–MS, Py–GC–MS, FTIR		Use of resinous exudates in Roman Britain, including coniferous resin, mastic/terebinth resin from the Mediterranean and frankincense from southern Arabia or eastern Africa (Brettell et al., 2014 , Brettell et al., 2015). Pitch from the wreck of King Henry VIII's flagship (1509–1545CE), the Mary Rose, identified as Stockholm tar, a good-quality pine tar obtained from the destructive distillation of <i>Pinus sylvestris</i> (Evershed et al., 1985). Detection of copal from East Africa and frankincense (produced locally) at the medieval port of Sharma (Yemen), informing about trade routes (Regert et al., 2008).

	Fermented beverages	Bacteriohopanes Other biomarkers controversial (tartaric acid, syringic acids, calcium oxalate, ergosterol) because analyses of few potsherds, non-specificity of analytical techniques, ubiquitous compounds highly labile or soluble in water.	Hopanoids characteristic of bacteria <i>Zymomonas mobilis</i> responsible for sap fermentation in pulque. Approach could be used for the detection of other bacterially-fermented alcoholic beverages such as palm wine, beer, cider, perry, and a wide range of other plant sap and fruit derived beverages.	GC-MS (SIM)		Detection of specific bacteriohopanoid distributions in amphorae from Teotihuacan (150 BCE to 650CE) provided evidence for the production of pulque (fermented sap of <i>Agavaceae</i> plants; Correa-Ascencio et al., 2014). Pine resin biomarkers for waterproofing detected in the same vessels.
Bitumen	Fossil organic matter	Hydrocarbons, steranes, terpanes.	Biomarker ratios, δD and $\delta^{13}C$ values can provenance bitumen to area of geographic origin.	GC-MS (SIM), GC-C-IRMS	Petrography	Identification of bitumen, mainly from the Dead Sea, often mixed with fat, conifer resin and beeswax, used in mummification practices by Egyptian embalmers between 1000 BCE and 400 CE (Connan, 1999). Combed ware pottery vessels from Ancient Greece used as beehives (Evershed et al., 2003). Beeswax mixed with tallow to make candles at 12th century Fountain Abbey, Yorkshire (Frith et al., 2004). Beeswax employed as a lamp illuminant in Minoan Crete (Evershed et al., 1997b). Beeswax used as a waterproofing agent for 5th mil. BCE collared flasks (Salque et al., 2013) and more generally widely used across Europe during the Neolithic, acting as an ecological indicator for the presence of the honeybee (Roffet-Salque et al., 2015). Beeswax used as widespread component of balms for mummification in Pharaonic and Graeco-Roman Egypt (Buckley and Evershed, 2001 ; Clark et al., 2013).
Insect	Waxes e.g. beeswax	Odd carbon numbered <i>n</i> -alkanes (C_{21} – C_{33}), even-numbered free fatty acids (C_{22} – C_{30}) and long-chain palmitate esters (C_{40} – C_{52}).	Presence of wax esters confirms beeswax.	GC-MS		

1968). Lipids extracted from archaeological pottery can be considered a reliable source of carbon based on two main properties: (i) they have fast metabolic turnovers and thus young ages at the time of deposition, and (ii) they are likely to be largely indigenous to the ancient pottery vessels because of their relative immobility and hydrophobicity in the burial environment (soil; Stott et al., 2003). The direct dating of lipids has developed from bulk analysis to the isolation of single compounds for radiocarbon dating (Stott et al., 2001, 2003). This approach limits the sources of exogenous contamination, thus minimising biased results. However, radiocarbon dating on lipids is highly challenging as it requires the isolation and purification of single compounds extracted from complex matrices (Stott et al., 2001). The targeted compounds from lipid residues are the C_{16:0} and C_{18:0} fatty acids from animal fats, as they are both the most common and abundant compounds identified in archaeological pots. Such direct dating of lipids residues has been proven possible and promising, since its first application more than 10 years ago on lipids extracted from sherds from the dendrochronologically dated Neolithic Sweet Track in Britain constructed between the winter of 3807/6 BCE and the spring of 3806 BCE (Berstan et al., 2008; Stott et al., 2001, 2003). With the advent of high precision accelerator mass spectrometry (AMS) for radiocarbon dating, a minimum quantity of around 20 µg of carbon is sufficient to produce a date. However, in practice 200 µg of carbon are needed to obtain a precise date with a reasonable error. It is noteworthy that concentrations of lipids above 200 µg per gram of sherd are routinely detected in archaeological sherds (Stott et al., 2001).

Every single sherd excavated from an archaeological site is thus a potential source of carbon for dating pottery use and associated events. This means the production of radiocarbon dates for lipids could further refine chronologies, being used to test established pottery typologies and also being linked directly to the commodities processed in the vessels during their use, affording absolute dates for addressing archaeological questions relating to dietary and subsistence practices. Finally, direct dating of lipids preserved in pots provides a unique opportunity for reliably dating archaeological sites where organic material such as charcoal or bones are absent from the archaeological record.

7. Conclusion

The characterisation of amorphous and/or invisible organic remains by analytical methods, such as chromatographic, spectrometric and isotopic techniques, has contributed significantly to a wide range of archaeological questions (Table 1). Firstly, organic residue analysis has considerably enhanced our understanding of the technologies involved in the production, repair and use of ancient ceramics. Lipid residues have provided valuable information about pottery function, for example by identifying the processing of commodities such as ruminant and non-ruminant carcass fats, dairy products, aquatic resources, plant oils and waxes (Table 2). This has increased our understanding of ancient diet and foodways and provided insights into herding strategies and early agricultural practices. However, it is important to note that compounds allowing an unambiguous interpretation are relatively rare, as to act as biomarkers the targeted compounds need to be specific and stable over archaeological timescales (Philp and Oung, 1988). Secure identification can only be performed on a narrow range of foodstuffs and organic materials, precluding the reconstruction of recipes. In conjunction with archaeozoological and archaeobotanical studies, patterns of diet and subsistence can be identified across temporal and spatial scales. It also needs to be mentioned that the analysis and subsequent interpretation of organic residues should always be applied within the context of the archaeology and palaeoecology of the settlement, region and/or period from which they derive. Organic residue information must be integrated with other lines of archaeological evidence, such as faunal and archaeobotanical remains, in order to provide meaningful answers to research questions. Furthermore, compound-specific $\delta^{13}\text{C}$ values of animal fats and plant waxes from organic residues in pottery can provide spatiotemporal isoscapes,

reflecting the ecological conditions driving the balance of C₃/C₄ vegetation (West et al., 2010). These insights into local ecologies can help us understand the relationships between people and their environment, and could potentially be used to assess the timing and impact of climatic events. The identification of exotic goods in secure archaeological contexts provides information about resource exploitation/acquisition and ancient trade routes and networks. Finally, over the next few years, the anticipated development of new methods for the direct dating of lipids preserved in archaeological artefacts will provide a new dating material from archaeological sites, opening up new possibilities for the refinement of pottery typologies and, importantly, the production of highly accurate chronologies.

The lipid residue approach is a powerful tool for archaeological investigations. The integration of transdisciplinary findings has already proven to be very productive, increasing our knowledge about ancient lifeways at house, site, regional and even continental scales. Further studies will certainly provide hitherto unseen insights into the past.

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